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HYDROCHEMICAL SOLITON: EXPERIMENTAL STUDY OF A CHEMICAL WAVE COUPLED WITH MARANGONI INSTABILITY IN BELOUSOV-ZHABOTINSKY REACTION

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It has been known the existence of the Big Wave (BW), which is a peculiar chemical wave, in a quasi 2-dimensional shallow layer of unstirred excitable Belousov-Zhabotinsky (BZ) reaction[1]. The specific feature of BW is the fact that it causes a single large solitary wave on the solution surface[1-3]. Therefore BW may belong to the category of the so-called "dissipative soliton"[3]. It has been made clear by some experimental studies that the physical mechanism of BW is due to the hydrodynamic instability coupled with chemical reactions[2-5].

BIG WAVE (HYDROCHEMICAL SOLITON)

The BZ reaction, one of the typical oscillatory chemical reactions, has been well studied as a representative example of dissipative structures. The spatio-temporal oscillations such as spiral waves or target patterns are observed in quasi 2-dimensional unstirred batch reactors of the BZ solution layers.

In the case of excitable BZ solutions, BWs can generate under proper conditions. Figure 1 shows a spatial pattern of BW in a petri dish (diameter: 85mm). This wave is quite different from the normal reaction-diffusion waves on the following five points[1-5].

- (1) The flow exists in bulk and on surface of the solution layer.
- (2) It causes large surface deformation (nearly 5 μ m).
- (3) It propagates acceleratingly with high velocity.
- (4) The layer depth influences propagating properties.
- (5) It shows soliton-like behavior.

This wave is also called "Hydrochemical Soliton" from the reason (5). It is concerned with large change of enthalpy due to the chemical reaction and conspicuous gradient of surface tension of the layer[2].

Namely, it is a problem of the Benard-Marangoni instability coupled with the chemical reaction[2-3].

We can regard BW as "dissipative soliton" in some respects of its qualitative properties. Solitons in dissipative systems are most up-to-date topics, in contrast with solitons in conservative systems.

Figure 2 shows a temporal development of the surface deformation caused by BW, which was observed by an optical technique(Mach-Zehnder interferometry)[2]. We can see an accelerating propagation of a pulsive deformation.

In our previous experiments we observed the collision of two individual BWs. In this case, the two waves always vanished when they collided with each other. This phenomenon is due to the nature of excitable medium of BZ reaction, and is quite different from normal solitons.

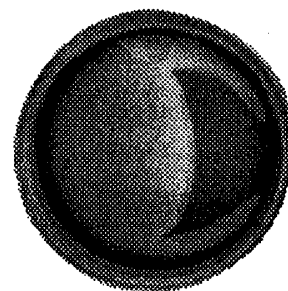


Fig.1 Propagation of BW in petri dish (2min after triggering). The wave propagates toward the right with acceleration.

MARANGONI INSTABILITY

As we already have described above, BW is a problem of coupling of the Benard-Marangoni instability and the chemical reaction[2].

It is investigated in terms of hydrodynamic instability that how the acceleration of BW depends on the depth of the solution layer[4,5]. The result is shown in Fig.3. This shows that when the depth is shallower than the critical value (near 1.4mm) the wave propagates with positive acceleration, while the wave loses its velocity and proper nature as the BW when the depth is deeper than that value.

This observation is consistent with the expected interpretation from the Benard-Marangoni instability[6-8]. In general, the Marangoni effect is more dominant in the shallower layer, while the Benard instability becomes to be more dominant in the opposite case[4-8].

The surface wave induced by the Marangoni instability is known as a wave which has the large wavelength(\sim cm) and causes large surface deformations in simple liquid systems[6-8]. The BW in BZ reaction would correspond to it in the points of their similarity[4,5].

We have described some properties of BW and the coupling of the hydrodynamic instability with the chemical reaction. We think BW as one of examples of the dissipative soliton. Nevertheless, the physical mechanism of the accelerating propagation has not been solved thoroughly. It must be necessary to investigate dynamics of the flow experimentally. Measurements of velocity distribution of the flow are now in progress.

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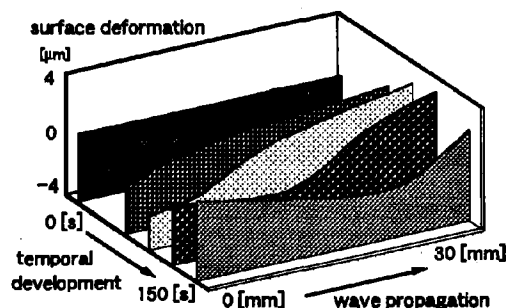


Fig.2 Temporal development of surface deformation caused by BW.

Several cross sections of the layer in time series are shown. The height 0 μm means that of initial stage.

A well which depth is about 2 μm propagates toward right with acceleration. The layer depth is 1.15mm.

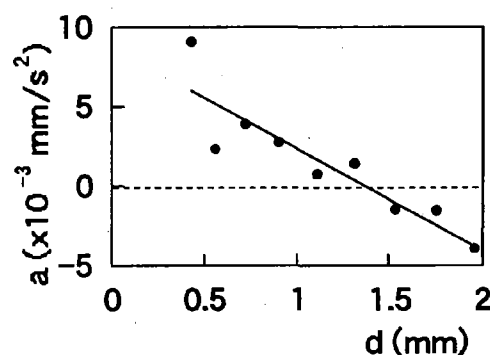


Fig.3 Depth (d) dependence of BW's acceleration (a) in a tilted petri dish[5].

The wave propagates to deeper region. The sign of a changes from positive to negative nearly at 1.4mm.